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The hyperfine structure of atoms informs us various static characteristics of nuclei, particularly for electro-magnetic moments and their distributions. We have been developing an experimental method to perform laser-microwave double-resonance spectroscopy for the hyperfine structure of Be and Ca isotopes, including unstable nuclei. The purpose and the status of the experiments are described.

1. Introduction

An experimental project to study unstable nuclei through hyperfine interactions using ion-traps has been proposed for one of the applications of the E-arena facility, a radioactive nuclear beam facility, of the Japanese Hadron Project. Laser spectroscopy on trapped ions using a double-resonance technique has great advantages in terms of precision and sensitivity. The ultra-high precision of $Q = 10^{13}$ in laser spectroscopy for a single ion has already been demonstrated in a study of frequency standard. Such high precision measurements enable us to study a new kind of physical quantity in nuclear physics. The hyperfine coupling constant A of the M1 term in the atomic hyperfine interaction shows a weak but finite isotope dependence called hyperfine anomaly, the main part of which stems from the finite size of the magnetization distribution in the nucleus (Bohr-Weisskopf effect) [?, ?]. This effect affords unique and interesting probe for valence neutron distribution in a nucleus, particularly in a so called neutron halo nucleus such as ^{11}Be . In contrast, the charge distribution is only sensitive to the proton distribution.

In this paper we describe the present status of the development for the experiment as well as proposed goals in physics.

2. Hyperfine structure and nuclear physics

2.1. Hyperfine interaction

Studies of the hyperfine structure of atoms have remarkably contributed to nuclear physics since early days. The ground state nuclear spins, magnetic dipole moments (μ_I) and electric quadrupole moments (Q_s) of many nuclei including unstable ones have been determined through measurements of the number of the hyperfine splittings and the magnitude of the splittings. Furthermore, the isotope shift (IS) due to the E0 hyperfine interaction, which appears in an optical transition between a certain state and an S state, affords useful probe for mean-square charge-radii of nuclei. It has been applied to long chains of isotopes and showed us fruitful phenomena such as the shell effect or the deformation effect etc.

The hyperfine interaction energies in an isolated atom is given as

$$W_{\text{hfi}} = A \left\{ \begin{smallmatrix} F & J & J \\ 1 & I & I \end{smallmatrix} \right\} + B \left\{ \begin{smallmatrix} F & J & J \\ 2 & I & I \end{smallmatrix} \right\} + C \left\{ \begin{smallmatrix} F & J & J \\ 3 & I & I \end{smallmatrix} \right\} + D \left\{ \begin{smallmatrix} F & J & J \\ 4 & I & I \end{smallmatrix} \right\} + \dots$$

where $\vec{F} = \vec{I} + \vec{J}$, I, J are the nuclear spin and electronic angular momentum quantum numbers, respectively, and A, B, C the hyperfine coupling constants. The relation of the nuclear moments and the hyperfine coupling constants which are classified by the multipolarity of the interaction is summarized in Table 1. The coupling constants A, B, C are proportional to the corresponding nuclear moments, μ_I, Q_s, Ω (magnetic octopole moment), respectively. The proportional coefficients are the hyperfine field strength of corresponding multipolarity at the nucleus. We can determine the nuclear moment from the coupling constant if the strength is obtained experimentally or theoretically. Since it is often possible to assume the field strength is constant among isotopes and the nucleus is a point, one can determine the nuclear moments in a certain accuracy with reference to the experimental data of a stable isotope. The high order moments, such as Ω (M3) or electric hexadecapole moment (E4), are the new criteria for nuclear physics, it ought to be opened by ultra-high precision spectroscopy for trapped ions. The precision for the hyperfine splittings measured by laser-microwave double resonance spectroscopy is often possible to resolve the high order coupling constants.

Analogous to the fact that the IS(E0) is due to the finite charge-distribution of the nucleus, the finite distribution of the nuclear magnetization causes another isotope effect

Table 1: HFS constants and nuclear moments.

<i>term</i>	<i>I, J</i>	<i>const.</i>	<i>moment</i>	<i>isotope effect</i>	
E0	≥ 0	-	$\langle r^2 \rangle eZ$	IS	(charge distribution)
M1	$\geq 1/2$	A	μ_I	B-W*	(magnetization distribution)
E2	≥ 1	B	Q_s		
M3	$\geq 3/2$	C	Ω		

* Bohr-Weisskopf effect[1, 2]

in M1 term, which is the so called differential hyperfine anomaly. The M1 coupling constant A is proportional to the magnetic field produced at nucleus by the orbital electrons, and to the nuclear magnetization. For a point nucleus this is taken to be the nuclear g value ($g = \mu_I/I\mu_N$, μ_N : nuclear magneton). Thus for an actual nucleus

$$A = A_{\text{point}}(1 + \varepsilon),$$

which defines the hyperfine anomaly ε , referred to as the Bohr-Weisskopf effect. Although direct determination of ε of an electronic atom, which is usually less than a few percent, is quite difficult, the differential hyperfine anomaly for a pair of isotopes

$${}^1\Delta^2 = \frac{A_1/g_1}{A_2/g_2} - 1 \simeq \varepsilon_1 - \varepsilon_2$$

can be obtained by precision measurement of A and nuclear g value for each isotope of the pair.

The Bohr-Weisskopf effect ε was evaluated in a microscopic theory with a factorization approximation by Fujita and Arima [3] as

$$\varepsilon = \langle |N(R) - K(R)| \rangle_{\text{VM}} + \frac{1}{\mu_I} \langle |K(R)| \rangle_{\text{VM}} \langle II | \sum_i g_s^{(i)} \Sigma_i^{(1)} | II \rangle,$$

where

$$N(R) \equiv \int_0^{R_i} FG dr / \int_0^\infty FG dr,$$

$$K(R) \equiv \int_0^{R_i} FG \left(\frac{r}{R_i} \right)^3 dr / \int_0^\infty FG dr,$$

R and r are nuclear and electron coordinates, F and G the Dirac electron wave functions, $g_s^{(i)}$ and $\Sigma_i^{(1)}$ the spin g value and the spin asymmetry operator of i th nucleon, and $\langle \rangle_{\text{VM}}$ means an expectation value of valence nucleons.

The radially dependent terms $\langle |N(R)| \rangle_{\text{VM}}$ and $\langle |K(R)| \rangle_{\text{VM}}$ may give us a possibility of investigating the valence nucleon density distribution. As far as the proton density distribution is concerned, there are several experimental ways such as electron scattering, IS, etc. For the neutron distribution, one way is to measure the transverse term in electron scattering [4]. The hyperfine anomaly can be another way of investigating it. Fujita and Arima mentioned it for the case of muonic atoms, since the calculation of $N(R)$ and $K(R)$ is possible for the muonic atoms, on the other hand, it is very difficult for electronic atoms due to a very complicated screening effect. When unstable nuclei are concerned, however, neither electron scattering nor muonic X-rays has been applied up to now. It is expected that the complexity of the calculation of electronic wave function for very light atoms such as Be isotopes can be relaxed to some extent. It should be mentioned for a case of very light isotopes, ${}^6\text{Li}$ and ${}^7\text{Li}$, as an example [2]. McColm [5] precisely calculated the differential hyperfine anomaly

$${}^n\Delta^7 = 1 - \frac{A_7/g_7}{A_6/g_6} \left(\frac{M_6}{M_7} \right)^3,$$

where M is the reduced mass. The experimental value of ${}^6\Delta^7 = 1.071(8) \cdot 10^{-4}$ [6] was compared with the theoretical calculation. If the experimental value is used to

determine the p-orbital nucleon radial parameter R_p , a value of $0.17 \cdot 10^{-4} \cdot a_0$ is obtained which is not inconsistent with that obtained from electron scattering experiment ($R_p = 0.20 \cdot 10^{-4} \cdot a_0$).

2.2. Present goals in physics

We take Be isotopes for the first object, particularly for measurement of the magnetic hyperfine splitting constant A of ^7Be in order to determine the nuclear magnetic moment with a certain accuracy. In spite of the importance of the magnetic moment of ^7Be , which has been discussed with a simple cluster model consisting of ^4He and ^3He , it is still unknown mainly due to the experimental difficulty. The ^7Be nucleus decays into ^7Li with a half-life of 53.3 d by the electron capture process (no β -rays). The 478 keV γ -rays are emitted followed by the decay, however, the spin of the initial state of the γ -transition fed by the β -decay is 1/2, unfortunately. Therefore neither β -NMR nor γ -NMR method can be applied to measure the magnetic moment. It is, however, very suitable for the optical measurement, in particular for the precision spectroscopy of trapped ions. The highly accurate nuclear magnetic moment of ^7Be will be determined from the measurement of the splittings of hyperfine magnetic sublevels in an external strong homogeneous magnetic field using a superconducting Helmholtz magnet, which is being under tested at off-line in the present project.

Following the ^7Be experiments, we will go to a very short-lived isotope, ^{11}Be ($T_{1/2} = 13.8$ s). Not only the magnetic moment of the ^{11}Be but also the hyperfine anomaly of the ^{11}Be is considered to be a very important quantity for nuclear physics. ^{11}Be is known as a neutron halo nucleus; a valence neutron is loosely bounded to the core and radially extended [7]. The hyperfine anomaly of ^{11}Be is expected to be very large due to this particular radial behavior. Comparison of $^7\Delta^9$ and $^9\Delta^{11}$ will let us investigate the difference of the valence neutron distribution in such neutron halo nucleus by a pure spectroscopic method.

A chain of odd Ca isotopes are also in our experimental plan. Direct determination of the magnetic moment for Ca isotope is rather complicated due to a presence of metastable states in low lying atomic levels, unlike Be isotopes. It is, however, most of the magnetic moments of Ca isotopes have been measured in sufficiently high accuracy. We can investigate the hyperfine anomaly of Ca isotopes by measuring the magnetic hyperfine constants A .

3. Experimental status

Our experimental plan is based on the development of two basic techniques: 1) trapping of unstable nuclear ions from an isotope separator and 2) precision spectroscopy for trapped ions using the laser-microwave double resonance. For the former, a new kind of linear RF-trap has been developed. A low energy continuous ion beam from an ion-guide gas cell was accumulated in the linear trap by a cooling of frequent collision with He buffer gas of $0.1 \sim 1$ Pa. The bunched ions were extracted and guided to a subsequent vacuum chamber through skimmers. For the latter, laser cooling is an essential technique to perform precision measurements for unstable nuclear ions which are usually obtained in very few amount. In an off-line ion-trap at ILS/UEC, a single

ion and an Wigner crystal consisting of a few ions of ^{40}Ca was observed by a photon counting camera as a result of laser cooling down to a very low temperature of 0.5 K.

In the following some details of these development are reported.

3.1. Trapping of ions in a continuous beam

In general, unstable nuclei are produced by bombardment of a target with accelerated particles and separated by an isotope separator on-line (ISOL). The mass separated unstable nuclear ions are usually obtained in an energetic ($\sim\text{keV/u}$ for an ion-source type separator and $\sim\text{MeV/u}$ for a recoil type separator) continuous beam. Collection of such ions into traps is an important practical concern in precision spectroscopy experiments for unstable nuclei. Manipulation of such ion beams involves modifying the phase-space volume as well as the absolute velocity. The gas of various pressure at several stage of the trapping instruments plays important roles in cooling (decreasing in phase-space volume) and in degrading (decreasing in absolute velocity).

We have been developing a new efficient trapping method for unstable nuclear ions from a gas-filled recoil separator (GARIS) using an ion-guide, a SPIG (sextupole RF ion beam guide) and a linear trap. It has been tested with an off-line ion-guide apparatus up to now.

The ion-guide is an ion collection system which has originally been developed in Jyväskylä for a sort of ion-source of an ISOL [8]. A reasonably large fraction of recoiled out reaction product from the target is stopped in the He gas cell (~ 10 kPa) and kept in a singly charged state. Then such thermalized ions are guided by gas flow toward the exit hole (< 1 mm ϕ) and extracted by an electric field applied between the cell and a skimmer while the gas is evacuated by a roots pump. The combination of the GARIS and the ion-guide has been tested at INS [9].

The SPIG has been developed to guide ions extracted from the ion-guide gas cell without any acceleration, and to bring them to a high-vacuum region through skimmers with high efficiency ($> 90\%$) and a small energy spread [10, 11]. A cylindrical aligned six rods, on which RF potentials are applied, forms a guiding field similar to an optical fiber. Ions coming out from the gas cell are smoothly guided by this radial confinement field without an extraction field and the gas is efficiently evacuated through the wide apertures between the rods.

We have developed a new style of RF linear-trap using the SPIG in which ions in a continuous ion beam can be accumulated. Three rings placed outside the SPIG rods with a coaxial configuration, which are kept at different static potentials, form the linear trap (Fig. 1). Due to a penetration of the field generated by the outer rings, the average potential at the center of the SPIG can be slightly shifted from the mean potential of the SPIG rods. By adjusting the voltage of the three rings, an axial trapping potential can be produced. Since the trapping region is filled with He gas of ~ 1 Pa, in which the mean-free-path of ions is on the order of 1 cm, ions from upstream can be accumulated by gas cooling.

We have searched for an optimum condition for the trapping of ions. A fraction of the low-energy continuous ion beam in the SPIG was trapped in this linear trap and extracted as a bunch. A typical signal of trapped ions detected by a Faraday cup at the end of the SPIG is illustrated by a sequence diagram in Fig. 2. In the ‘Thru’ period,

Figure 1: Schematic layout of the new RF linear trap with the ion-guide and the SPIG. Ions are produced in the He gas cell by an arc discharge, and guided to the exit nozzle by a He gas flow. An RF voltage of ~ 100 V_{pp} (6.7 MHz) is supplied to the SPIG rods, and forms a sextupole RF field to create a radial confinement field. Three ring electrodes (Ru, Rc and Rd) placed outside the SPIG are connected to fast high-voltage amplifiers by which the axial trapping field is controlled. The SPIG rods pierce through three skimmers (the last one is not drawn); the pressure distributions (from upstream) are $1.1 \cdot 10^1$, $1.2 \cdot 10^{-3}$ and $1.5 \cdot 10^{-4}$ Pa when the gas cell is 25 kPa. The actual dimensions are: SPIG rods, 30 cm in total length, 0.5 mm in diameter, 2.8 mm p.c.d.; skimmers, 2.5 mm in inner diameter, 20 mm in length; rings, 4.4 mm in inner diameter, 5 mm (Ru, Rd) and 10 mm (Rc) in length with 0.5 mm separation gaps.

Figure 2: Typical signal of trapped ions and time sequence of the applied voltages to the three-ring electrodes (Ru, Rc and Rd). Ions in a continuous ion beam of 2 nA were accumulated for 9 ms (Acc) and held for 0.6 ms (H), then, extracted (Ext) as a bunched ion beam (see text).

Figure 3: Trapping efficiency and capacity (left: a), and life-time (right: b). a) Trapped ion peak current (thick line) and trapping efficiency (thin line) for different ion-source intensity ($\diamond$: 8.5 nA, $\square$: 5 nA, $\circ$: 2 nA) are plotted as a function of accumulation period. He gas cell pressure was 25 kPa. b) Trapping life-time. Trapped peak current normalized by 'Thru' DC current for different He cell pressure ($\diamond$: 25 kPa, $\square$: 20 kPa, $\circ$: 16 kPa) are plotted as a function of 'Hold' period. Vacuum at the trap region when He gas cell pressure was 16, 20 and 25 kPa was 0.35, 0.6 and 1.3 Pa, respectively.

all of the rings were grounded, and a DC current of 2 nA was detected. In the 'Acc' (accumulation) period, the downstream ring (Rd) was turned to 70 V and the central ring (Rc) was to -5 V in order to produce a potential barrier and well, respectively. In the 'Hold' period, the upstream ring (Ru) was also raised to 100 V in order to stop the supply of ions from the upstream gas cell. In the 'Ext' (extraction) period, Rd was grounded and Rc and Ru were raised to 80 V and 150 V, respectively, in order to extract the ions in the potential well as a bunch. A short pulse at the beginning of 'Ext' is the signal of bunched ions accumulated for 9 ms and held for 0.6 ms. The number of ions in the pulse was estimated to be $7 \cdot 10^6$, which corresponds to a 7% fraction of the ions supplied in the accumulation period.

Some performances were measured. By varying the accumulation period, the accumulation capacity and trapping efficiency were measured (Fig. 3 a)). Saturation at a peak current of 15 nA was observed. Because the saturated peak currents were independent of the intensity of the supplied ion current ('Thru' DC current), the saturation seems to be due to the capacity of the trap, which is limited by the space-charge effect. Based on the same measurements, the trapping efficiency, which is here defined as the ratio of the total charge of the trapped ion signal to the number of applied ion, which is a product of accumulation period and the 'Thru' DC current, was estimated. The trapping lifetime was also measured by varying the 'Hold' period after a certain fixed 'Acc' period (Fig. 3 b)).

The efficiency was found to be decreasing upon increasing the accumulation period. Only a fraction of ions which survived in competition with some ion-loss processes was trapped. The origins of the ion-loss process are supposed to be collisions and charge exchange with some heavy atoms or molecules, and RF heating by the trapping field. In the present case, the latter is considered to be dominant. Since the RF heating rate strongly depends on the space-charge effect, it is sensitive to the intensity of the supplied current. This is also supported by lifetime measurements. A longer lifetime

was obtained with a higher buffer gas pressure; it means that a higher cooling rate, which accompanies more impurities, though, is important to overcome RF heating.

Although the capacity of $7 \cdot 10^6$ ions is a quite large number for the trapping of unstable nuclear ions, it can be increased by using a longer central-ring electrode, if one needs more capacity. The trapping lifetime of 90 ms seems to be longer than that estimated from the poor background vacuum of 10^{-2} Pa (without He gas) in the trap region. We need to increase the life-time to the order of 1 second in order to accumulate ions in a very low-intensity beam. It would be achieved by further improvements, such as to realize a higher background vacuum with a higher density buffer gas of high purity He, and to add one or more trapping region with a variable length in which ions are accumulated in large volume and then compressed.

Once ions are bunched in the linear trap, it would be easy to transfer them to a subsequent ion traps, which are at a UHV chamber. The present linear trap will be used in combination with a new high-resolution mass separator at INS, as well as with the GARIS-type separator. The new ISOL was designed not only for the radioactive beam acceleration but also for the ion-trapping. Since the entire mass separator can be electrically floating on a high voltage platform (floating-ISOL), after deceleration, a very low energy ion beam can be obtained at the ground potential apparatus. For this on-line trapping, a new mechanism at the front end of this linear-trap must be developed where an additional differential pumping system and a pulsed gas cooling mechanism must be elaborated.

3.2. Laser cooling and spectroscopy

The off-line trap experiment was carried out mainly aiming at a feasibility study of precision laser-microwave spectroscopy for short-lived nuclear ions. In general, the ion-trapping technique is not considered to be suitable for a short-lived nucleus, because one of the advantages of this technique is to provide a long confinement time in an isolated space, which is not very effective for a short-lived nucleus. If, however, a single-ion spectroscopy for unstable nuclei is realized, this drawback would somehow be relaxed due to the following reason. Suppose that we prepare a thousand ions at first; we could

Figure 4: Schematic diagram of the off-line laser experimental apparatus.

Figure 5: Image of three $^{40}\text{Ca}^+$ ions aligned in a linear trap and a time spectrum showing a discrete decay of fluorescence intensity accompanied by decrease in the ion number.

then expect one ion even after a ten-times longer period of the half life of the nucleus.

We have tested the scheme at off-line using stable Ca ions. A schematic drawing of the experimental apparatus is shown in Fig. 4 and details of it are described elsewhere [12]. Figure 5 is an image of a crystal comprising three $^{40}\text{Ca}^+$ ions in an off-line quadrupole linear trap. It was observed using a two-dimensional photon-counting camera (Hamamatsu PIAS-T1). The intensity of the fluorescence from a single ion was about 200 cps at an UV laser power of $300\text{ }\mu\text{W}$. The single ion repeated the cycle of absorption and emission with a frequency of as high as 4 MHz, which was estimated by using a detection efficiency of 5×10^{-5} . The counting rate is sufficient to perform a high-precision spectroscopy experiment.

Prior to hfs spectroscopy experiments with a single laser-cooled ion, we tested a detection of a resonance in the kinetic motion of stored ions [13]. It is possible to increase the kinetic energy of the ions by applying an external perturbation field, if the frequency is resonant to the characteristic motion of the ions in the trap. The frequency of this characteristic motion, the so-called secular frequency, is

$$\omega_{\text{sec}} = \frac{1}{\sqrt{2}} \frac{eU}{m\omega r_0^2}.$$

where e, m are charge and mass of the ion, r_0 bore radius of the linear trap and U, ω amplitude and frequency of the RF. This resonance is detected by a decrease in the fluorescence intensity caused by the resonant heating of the ion motion. Figure 6 shows a typical spectrum of the secular resonance which was detected by a single $^{40}\text{Ca}^+$ ion. Since the secular frequency is inversely proportional to the mass of the ion, this is a kind of mass spectroscopy. Although the mass resolution in the present experiments was not very good for precise mass measurements, it showed the possibility of resonance detection with a single ion in our apparatus. It should be noted that some other small resonant peaks corresponding to ions of different mass were also observed. It was thought to be due to sympathetic heating (cooling) [14]. The single laser-cooled ion did play the role of a detector of resonant heating of other, non laser-cooled, ions. If one performs this experiment in a precision Penning trap, where the cyclotron frequency (eB/m) could be precisely measured [15], it may open a new way of precision mass spectroscopy for a wide variety nuclear ions, even though they cannot be laser cooled.

Figure 6: Secular resonant spectra detected by a laser cooled single $^{40}\text{Ca}^+$ ion. a) Narrow-band slow-scan (40 Hz/s) spectrum with a weak rf signal (30 mV). The asymmetry is explained by considering that since the cooling rate is not as fast as the heating rate, the drop (left side) is quickly caused by a resonant-heating effect, and the recovery (right side) is slow. The mass resolution was estimated to be $\sim 1/200$. b) Mass spectrum calibrated by $^{40}\text{Ca}^+$. It was taken by a wide-band rapid-scan (100 Hz/s) with stronger rf (50 mV). Some impurity ions of $A=39, 46, 48, 49$ and 50 were detected by the sympathetic heating effect. The signal after $A=38$ is the background where the Ca ion disappeared.

Figure 7: Laser cooling rate. a) Fluorescence intensity as a function of the time after a YAG laser shot by which ions are produced from metallic Ca. The intensity is suddenly increased and saturated within a certain cooling period, which is our definition of the cooling time (and reciprocal of the cooling rate). Several runs under the same condition (off-tuning of the UV laser was ~ -300 MHz) were plotted and from which a cooling time of 2.57(13) s was derived. b) Cooling rate as a function of the laser power. Since the rate seems to be proportional to the laser power until the S - P transition is saturated, and the coefficient was derived to be 20 Hz/mW, a quicker cooling time of more than one order of magnitude would be achieved by a mW class laser.

One more very important test concerning the feasibility study is to examine how quickly ions can be cooled. The cooling time (rate), which is here defined as a period between the production of ions and the sudden increase of fluorescence to a saturated count rate, was measured as a function of the laser power (Fig. 7). A cooling time of 2.6 seconds was obtained at 22 μ W of laser power, and the rate seems to be proportional to the laser power. The cooling time, then, can be decreased by increasing the laser power until saturation of the strong *S-P* atomic transition occurs. It should be mentioned that the Ca ions in those experiment were produced by laser ablation from a metallic Ca mounted in the trap: the initial kinetic energy is considered to be very high, on the order of 10 eV, which is much higher than that of ions from upstream linear trap (~ 0.8 eV) in the future on-line experiment.

For the future experiment for less abundant isotopes, such as ^{43}Ca , we have tested an implanted Ca source in a Be disk. Since mass separated pure isotopes can be provided easily in such implanted form. We used a Be disk in which $2 \cdot 10^{12}$ ^{40}Ca atoms were implanted within an area of about 50 mm². A sufficiently large amount of Ca ions were produced for many times from the same spot of the ablation laser shots. This result suggested that the method of laser ablation from an implanted source can be used also for some long-life radioisotopes such as ^7Be or $^{41,44,45,47}\text{Ca}$ as well as stable ones. The strength of radioactivity of 10^{12} atoms of ^7Be is as weak as 1.5 kBq.

4. Conclusion

Development of the techniques described are ready to be combined to perform precision spectroscopy of unstable nuclei at the new ISOL (isotope separator on-line) system of INS as well as at the GARIS. An off-line experiment for ^9Be has also started recently. A series of Be ($A=7,9,11$) and Ca (39-49) isotopes will soon be studied in our on-line ion-trap system.

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